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Preparation and Characterization of High Dielectric Constant Polymer Films

Nick Harrison
Jeff Szabo

Defence R&D Canada

Technical Memorandum
DRDC Atlantic TM 2002-159
September 2002

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Author

Jeff Szabo

Approved by

Richard Morchat

Head/ Emerging Materials

Approved for release by

Kirk Foster

DRP Chair

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Abstract

Electroactive polymers are an emerging class of actuating materials capable of generating large displacements and fast response times. Dielectric actuators operate on the principle of Maxwell stress and their response is highly dependent on the material's electrical and mechanical properties. Unfilled and composite films based on a thermoplastic polyurethane were prepared using solvent cast methods, and their dielectric properties were determined. Dielectric constants of up to 52.5 at 100 Hz were measured. The observed permittivities of the composites were consistent with the predictions of the Bruggeman model. Dielectric breakdown strength varied greatly from position to position on each film, reflecting the large statistical variation in flaw density over each film's area. For polyurethane/$BaTiO_3$ composite films, there was no significant change in breakdown strength with increasing barium titanate concentration.

Résumé

Les polymères électroactifs constituent une classe nouvelle de matériaux actionneurs à court temps de réponse capables de produire de grands déplacements. Les actionneurs diélectriques fonctionnent selon le principe des contraintes de Maxwell et leur réponse dépend étroitement des propriétés électriques et mécaniques du matériau. Des films non additionnés d'une matière de charge et des films multicouches faits d'un polyuréthane thermoplastique ont été préparés au moyen de méthodes de moulage en solvant, et leurs propriétés diélectriques ont été déterminées. On a mesuré des constantes diélectriques atteignant parfois 52,5 à 100 Hz. La permittivité observée pour les composites était conforme aux prévisions du modèle de Bruggeman. La résistance au claquage variait fortement d'un endroit à l'autre sur chaque film, illustrant ainsi la grande variation statistique de la concentration des défauts dans chacune des parties du film. Dans le cas des films composites faits de polyuréthane et de titanate de baryum ($BaTiO_3$), l'augmentation de la concentration en $BaTiO_3$ n'a pas entraîné de modification importante de la résistance au claquage.

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Executive summary

Background

A wide variety of actuators with a broad range of applications currently exist. For certain applications, however, many of these devices are unsuitable; the need for smaller, more efficient, versatile actuators has led to an increased interest in alternative actuation technologies. Dielectric polymer actuators are a promising class of materials offering high elongation combined with fast response. Although they have been in development for a number of years, there has been little effort to select or develop appropriate materials.

Principal Results

Elastomeric films were prepared by solvent casting a thermoplastic polyurethane with the additives barium titanate or poly(copper phthalocyanine). A drawdown applicator was used to create thin films. The measured dielectric constants of composite materials were consistent with the predictions of the Bruggeman model, suggesting that large gains in permittivity can only be achieved with high filler concentrations (>50% w/w). The addition of barium titanate up to a mass fraction of 0.4 had no significant effect on the breakdown strength of the polyurethane films.

Significance of Results

This study demonstrated that substantial increases in a film's permittivity are difficult to achieve unless the mass fraction of filler is exceptionally high. Additionally, it was found that fillers can be included in a film to increase the dielectric constant without reducing the breakdown strength.

Future Work

A suitable elastomer and appropriate fillers need to be developed or selected for use as an actuating material. Since the dielectric polymer actuators will operate at relatively high voltages, the high field permittivity of the films will be studied. Also, actuators will be fabricated and their response characterized. Finally, modeling will be carried out to relate a material's properties to actuator response.

Harrison, N. R., Szabo, J. P. 2002. Preparation and Characterization of High Dielectric Constant Polymer Films. DRDC Atlantic TM 2002-159. Defence R&D Canada – Atlantic.

Sommaire

Contexte

Il existe actuellement une vaste gamme d'actionneurs qui se prêtent à un large éventail d'applications. Toutefois, nombre d'entre eux ne conviennent pas à certaines applications; le besoin d'actionneurs plus petits, plus efficaces et polyvalents a entraîné un intérêt accru pour les autres technologies d'actionnement. Les polymères diélectriques actionneurs constituent une catégorie prometteuse de matériaux qui offrent un allongement élevé combiné à une réponse rapide. On travaille à leur mise au point depuis de nombreuses années, mais peu d'effort ont été orientés vers le choix ou la mise au point de matériaux adaptés.

Principaux résultats

Des films d'élastomère ont été préparés par moulage en solvant d'un polyuréthane thermoplastique, en utilisant comme additifs le titanate de baryum ou la poly(phtalocyanine de cuivre). On a utilisé un applicateur pour obtenir des films minces. Les constantes diélectriques des matériaux composites étaient conformes aux prévisions du modèle de Bruggeman, indiquant qu'une augmentation importante de la permittivité ne peut être obtenue qu'à des concentrations élevées de matière de charge (>50 % p/p). L'ajout de titanate de baryum jusqu'à l'obtention d'une fraction massique de 0,4 n'a pas eu d'effet important sur la résistance au claquage diélectrique des films de polyuréthane.

Importance des résultats

La présente étude a montré qu'il est difficile d'obtenir des augmentations considérables de permittivité sans une fraction massique de matière de charge exceptionnellement élevée. De plus, on a constaté que l'ajout de matières de charge à un film permet d'augmenter la constante diélectrique sans diminuer la résistance au claquage.

Travaux à venir

Il faudra mettre au point ou choisir un élastomère et des matières de charge appropriés qui seront utilisés comme matériaux actionneurs. Les polymères diélectriques actionneurs fonctionnant à des tensions relativement élevées, on étudiera la permittivité des films à un champ élevé. De plus, on fabriquera des actionneurs et l'on caractérisera leur réponse. Finalement, on procédera à des modélisations pour établir la relation entre les propriétés d'un matériau et la réponse de l'actionneur.

Harrison, N. R. et J.P. Szabo, 2002. Préparation et caractérisation de films polymères à constante diélectrique élevée. RDDC Atlantique TM 2002-159. Défense R & D Canada – Atlantique.

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1. Introduction

An actuator is a device that converts electrical energy into mechanical energy that can be used to do work. Electromagnetic motors and solenoids are common examples and are well suited for many applications. For other applications, however, several shortcomings are apparent, particularly when it comes to size requirements and versatility. In recent decades, the need for smaller, more efficient actuators has led to an increased interest in alternative actuation technologies. Piezoelectric materials offer fast response and large forces, but are limited to low strains (< 1%). Shape memory alloys produce large strains, but are slow to respond[1].

Members of a promising class of actuating materials, known as electroactive polymers (EAPs), are capable of generating large displacements and exhibit fast response times. These materials may be able to bridge the performance gap between piezoelectrics and shape memory allows. Dielectrics in particular have a number of desirable properties[2], and research to develop dielectric polymer actuators is ongoing[1,2,3,4,5,6]. As Leidner and Hacker mention[2], though, to date there has been little effort to select or develop appropriate materials for this application.

Therefore, the objective of this work was to formulate and characterize dielectric thin films as part of a three-year study led by DRDC Atlantic to develop dielectric polymer actuators for use in active vibration damping[7]. More specifically, the effect of additives on a film's permittivity and breakdown strength was investigated, and several methods of film preparation were explored.

2. Theory

Dielectric polymer actuators operate on the principle of Maxwell stress[1,7]. A thin, insulating, elastomeric film is positioned between two compliant electrodes across which a potential difference is applied. The resulting positive and negative charges on opposite electrodes attract, compressing the film in the thickness direction and expanding its area as depicted in Figure 1. The actuator is essentially a parallel plate capacitor with a dielectric material inserted between the plates. Using this model, one can derive expressions for the static elongation and stress exerted by the actuator[7]:

$$\varepsilon = \nu\kappa\varepsilon_0 V^2 / Ed^2 \quad (1)$$

$$\sigma = \nu\kappa\varepsilon_0 V^2 / d^2 \quad (2)$$

where ε is the elongation normal to the film's thickness, ν is Poisson's ratio, κ is the relative permittivity (dielectric constant) of the film, ε_0 is the permittivity of a vacuum, V is the applied voltage, and E and d are the Young's modulus and thickness of the film, respectively. Notice that elongation and stress are proportional to the square of the applied voltage and inversely proportional to the square of the film's thickness. The theoretical maximum elongation and stress occur when the applied voltage is slightly

Figure 1. *Actuator operating principle*

less than the dielectric breakdown voltage, the potential at which the insulating film breaks down and conducts a current. The breakdown voltage itself is dependent upon the dielectric breakdown strength and thickness of the material in the following manner:

$$V_d = V_b d \tag{3}$$

where V_d is the dielectric breakdown voltage and V_b is the dielectric breakdown strength. Substituting equation (3) into equations (1) and (2) yields the maximum elongation and stress:

$$\varepsilon_{max} = \nu\kappa\varepsilon_0 V_b^2 / E \tag{4}$$

$$\sigma_{max} = \nu\kappa\varepsilon_0 V_b^2 \tag{5}$$

Therefore, elastomeric films designed to have inherently high permittivity and breakdown strength will be the most effective as actuator components.

3. Experimental

3.1 Film preparation

A thermoplastic polyurethane dissolved in DMF, sample 00-05-8227-90[8], was selected as a working matrix. Properties of the polymer are shown in Table 1 and Table 2. Films were initially prepared using a solvent cast method where the polymer solution was poured onto a level surface and allowed to spread out over a specified area. A polyethylene release layer was adhered to 1 mm thick aluminum plates, and silicone sealant borders were set around the perimeters in order to confine the solution to the desired dimensions. Once the solutions were cast, the plates were placed in an oven at 80°C for a minimum of 48 hours to remove most of the solvent. A 29 in. Hg vacuum was then applied for a minimum of 8 hrs to complete the drying process.

Filled polymer films were prepared using the same casting technique. Additives included poly(copper phthalocyanine) provided by Bodycote Ortech and several barium titanate powders (<2 micron, Aldrich; NanOxide™ HPB-1000, TPL Inc.). The poly(CuPc), supplied as a pressed disc, was ground with a mortar and pestle before it was added to the polyurethane solution. The $BaTiO_3$ was added directly. The resulting blends were hand-mixed using a glass rod for approximately 10 minutes, and films of varying filler concentration were cast for permittivity measurements.

Table 1. *Properties of polyurethane*

PROPERTY		VALUE
Dielectric Constant[1]		9
Dielectric Loss[1]		0.41
Solution	Solvent	DMF
	% w/w	40
	Viscosity	High
Film		Elastomeric solid

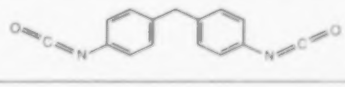
[1] at frequency = 100 Hz. See Figure 6.

To prepare films for dielectric breakdown strength measurements, it was necessary to use a different method than described above. When the fillers were added to the polyurethane solution, the viscosity increased significantly and the mixture could not flow to fill the proposed area. This made it impossible to prepare thin films. Furthermore, the films proved difficult to release from the polyethylene backing.

As a solution, a drawdown device (Gardco) was obtained. This simple tool (Figure 2) allows the user to prepare films of uniform thickness down to 25 μm by pouring the polymer solution and drawing the applicator over it. Samples for breakdown tests were not cast on the polyethylene backing but directly onto an aluminum plate, which served as one electrode in the breakdown strength apparatus. Consequently, there was no need to release the films and good contact between the electrodes and samples was assured.

The films were dried using a vacuum oven in the same manner as before. To minimize dust contamination, the dried samples were placed in a fumehood where they remained until tested.

Table 2. *Molecular composition of polyurethane*

REACTANT	MOLAR RATIO	MASS RATIO	STRUCTURE
poly(tetramethylene ether glycol) MW=1400	1	69.472	
2,2-dimethyl-1,3-propanediol (DMPD)	1	5.258	
4,4'-methylenebis(phenyl isocyanate) (MDI)	2	25.269	

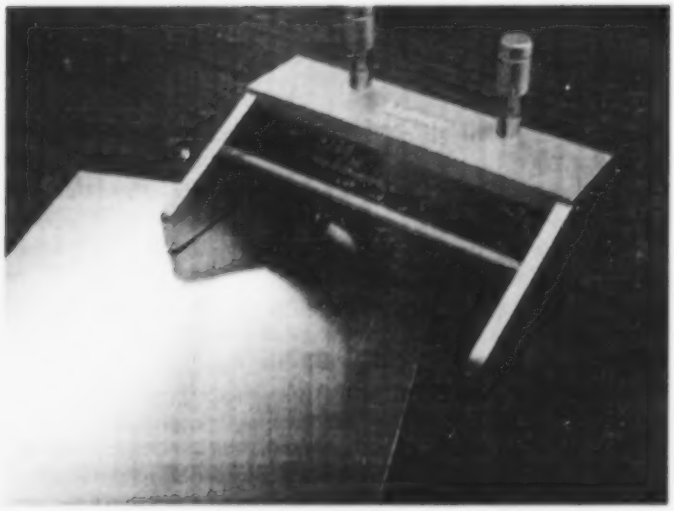


Figure 2. *Drawdown device*

3.2 Permittivity

Permittivity measurements were carried out using the DEA 2970 Dielectric Analyzer (TA Instruments) and accompanying Thermal Analysis software. Samples (1 x 1 in.) were cut and removed from the polyethylene release layer. On occasion, it was necessary to cool the film in a freezer (-10°C) prior to release to avoid stretching and tearing the sample. Film thickness ranged from 0.26 mm to 0.76 mm. Barium titanate composites tested for permittivity were made using < 2 micron barium titanate powder from Aldrich.

The experimental run performed by the DEA 2970 on each sample included a temperature equilibration at 20.00 °C followed by a frequency sweep from 0.1 Hz to 100 kHz. Permittivity and loss were reported as a function of frequency.

3.3 Breakdown Strength

3.3.1 Apparatus Design

A device for measuring the dielectric breakdown strength of polymer films was constructed according to the schematic diagram presented in Figure 3. Waveforms were produced using a function generator (Model 145, Wavetek) and amplified by the Trek Model 20/20C High-Voltage Power Amplifier. Voltage and current waveforms were captured using a two-channel oscilloscope (Model TDS 310, Tektronix). For safety purposes, all high voltage components were placed inside a polycarbonate test chamber. A magnetically actuated safety interlock switch (Allen-Bradley) was installed to turn off the high voltage should the test chamber door be opened during the

experiment (see Figure 4a). A Fluke 189 digital multimeter and high voltage probe measured the potential across the electrodes, indicating whether or not the high voltage was on. Collectively, these precautions ensured no person came into contact with electrically energized parts.

Figure 4b shows the high voltage electrode, a stainless steel cylinder of 50.8 mm diameter and 25.4 mm height with edges rounded to approximately 2 mm radius. The contacting surface was polished to a near mirror finish using 240 grit abrasive paper. The grounded electrode was a 200 mm x 200 mm x 1 mm aluminum plate (i.e. the same plate on which the film was cast).

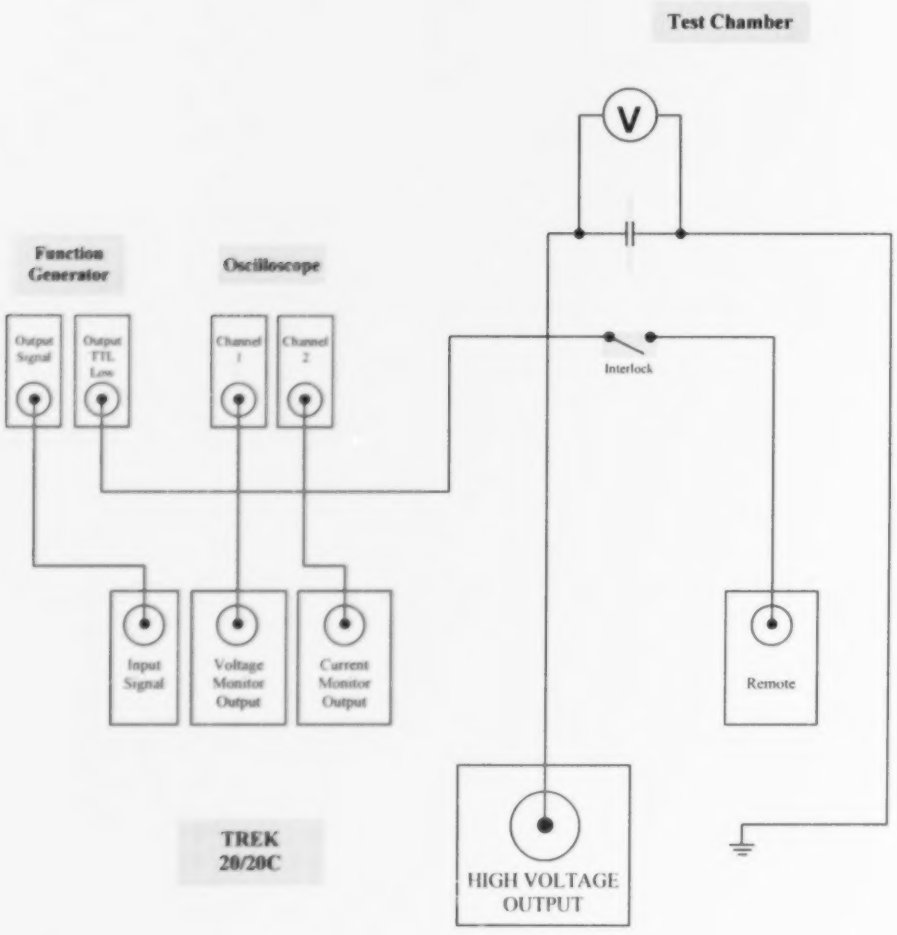
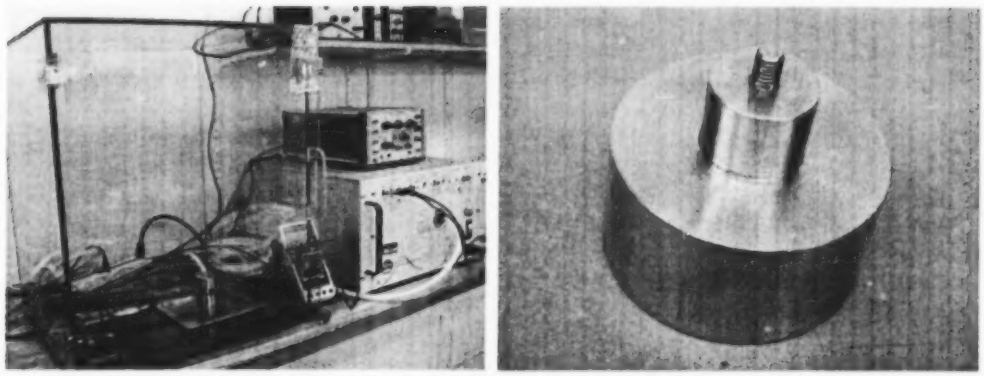


Figure 3. *Schematic diagram of dielectric breakdown strength test apparatus*



(a) (b)

Figure 4. *(a) Test chamber; (b) high voltage electrode*

3.3.2 Measurements

Breakdown strength tests were conducted at room temperature and humidity with air as the surrounding medium. The ASTM D 149 short-time test was used. During the voltage ramp, a sharp increase in current and a voltage drop to zero were indicative of breakdown (Figure 5 shows a typical breakdown curve). Following breakdown, the film thickness was measured with an electronic thickness gauge (Model ETG-2, Gardco). Finally, breakdown strength was calculated by dividing the breakdown voltage by the film thickness. $BaTiO_3$ composites tested for breakdown strength were made using NanOxide™ HPB-1000 from TPL Inc.

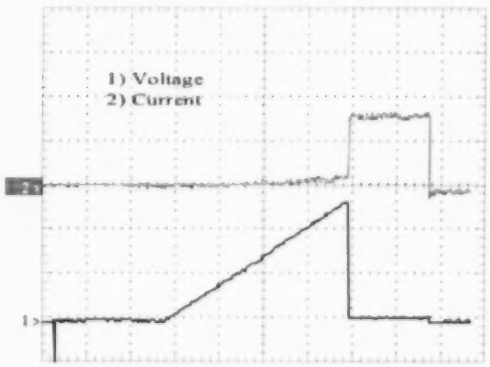


Figure 5. *Dielectric breakdown waveforms*

4. Results and Discussion

Preparation and handling of the thermoplastic polyurethane films was problematic. A significant issue concerned the addition and mixing of the additives—stirring by hand resulted in poor dispersion of the fillers, particularly the barium titanate. According to the suppliers, the average particle sizes of the barium titanate powder were 1.34 μm (Aldrich) and 50 nm (TPL Inc.) by FE-SEM analysis, yet visible clumps of filler 0.2 – 0.5 mm in diameter were present in the films. This inconsistency in particle

distribution has an adverse effect on a composite's homogeneity, which in turn affects its dielectric properties[9]. Use of a double planetary mixer may solve this problem. A second concern involved the removal of solvent once the polymer solution was cast. The vacuum oven method prevented sufficient airflow for unhindered evaporation of the DMF, which condensed on the vacuum door and was wiped off to remove it from the chamber. It is uncertain whether solvent remained in the films after drying, or what effect this residual solvent would have on film properties. Possible remedies include the use of a hotplate or convection oven, or avoiding the solvent casting method altogether. Finally, the physical and mechanical properties of the films were inadequate in terms of ease of handling. The tacky, adhesive-like nature of the polymer severely inhibited release from the polyethylene backing, and extreme care was needed to remove the films. Even when caution was taken, many samples were stretched and torn, most likely because the polymer chains are uncross-linked. Regardless of the material's shortcomings, the drawdown applicator was effective as a means of creating thin films.

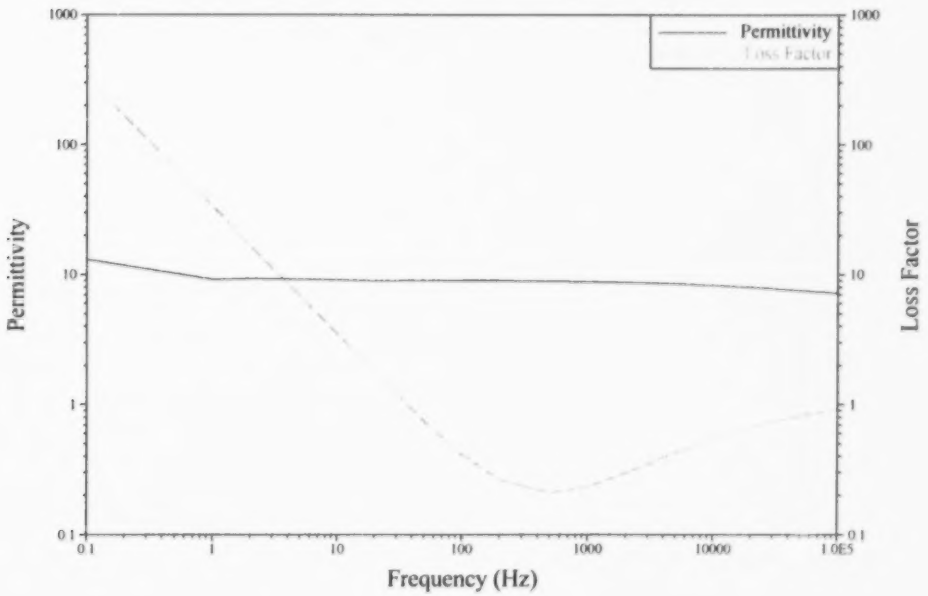


Figure 6. *Dielectric properties of polyurethane*

Dielectric properties of composite materials were shown to be dependent on filler concentration. The permittivity and loss of unfilled, $BaTiO_3$ filled, and poly(CuPc) filled polymer are shown in Figure 6 and Figure 7. Notice the dramatic increase in loss as the frequency drops below ~10-100 Hz. This behaviour results from the ionic conductivity of the material. To illustrate, the complex permittivity, ε^*, consists of real and imaginary parts:

$$\varepsilon^* = \varepsilon' - i\left(\varepsilon''_{dip} + \sigma/2\pi f\varepsilon_0\right) \tag{6}$$

where e′ is the permittivity, $i = \sqrt{-1}$, ε''_{dip} is the loss due to dipolar reorientation, σ is the ionic conductivity, f is the frequency, and ε_0 is the permittivity of free space. The real component represents the dielectric constant, whereas the imaginary component represents the loss factor. If the loss at frequencies below ~10-100 Hz is due entirely to ionic conductivity, a plot of loss factor versus inverse frequency should yield a straight line with slope $\sigma/2\pi\varepsilon_0$ passing through the origin for those frequencies. This is indeed the case, as shown in Figure 8. At this time, the identity of the ionic species is unknown; synthesis by-products and reaction impurities could be the source of low molecular weight, non-polymeric, mobile ions. Figure 7 shows another interesting yet unaccounted for loss characteristic concerning the addition of filler. Barium titanate composites tend to have lower loss than the unfilled polymer, while higher loss is seen in poly(CuPc) composites. This is rather unfortunate since the additive that increases the dielectric constant the most also increases the loss considerably. However, the modest increase in permittivity accompanied by a significant decrease in loss seen in $BaTiO_3$ composites suggests that the filler may prove useful in combination with other fillers.

The observed relationship between permittivity and filler concentration gives reasonable agreement with the Bruggeman model[10], which can be used to predict a composite's effective dielectric constant (ε_c) knowing the permittivity of the matrix (ε_m) and of the particle (ε_p), as well as the particle volume fraction (ϕ):

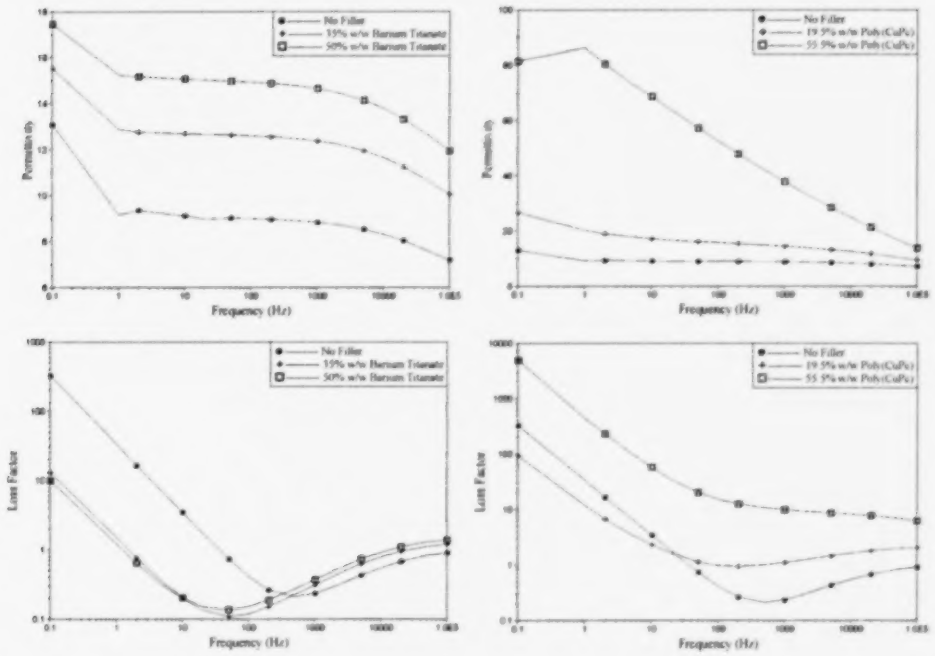


Figure 7. *Effect of (a) barium titanate and (b) poly(copper phthalocyanine) additives on permittivity and loss in a polyurethane film*

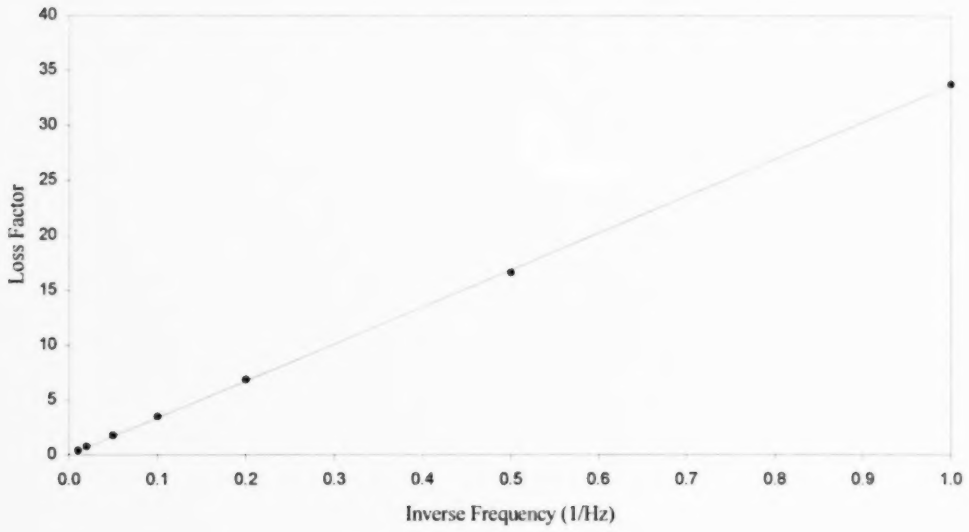


Figure 8. *Loss due to ionic conductivity*

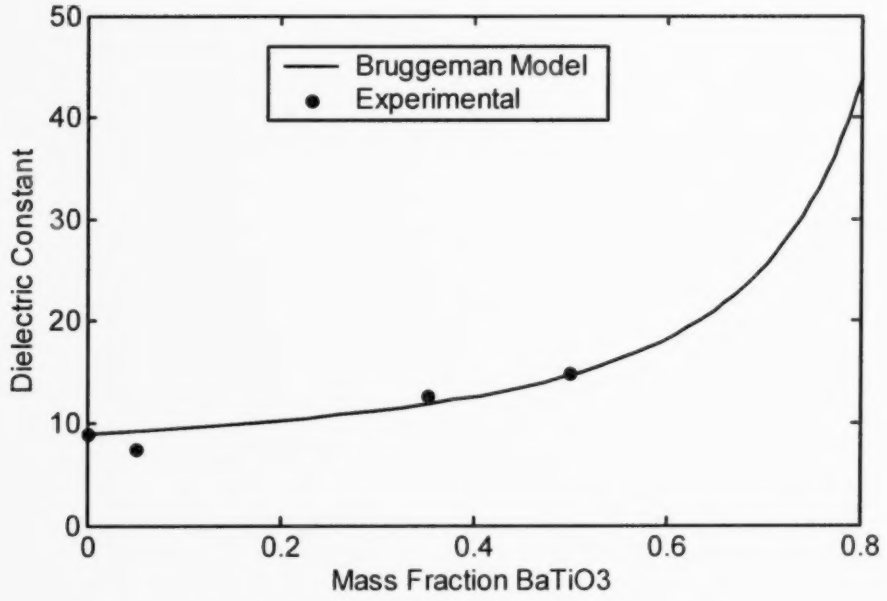


Figure 9. *Bruggeman model of polyurethane/barium titanate composite*

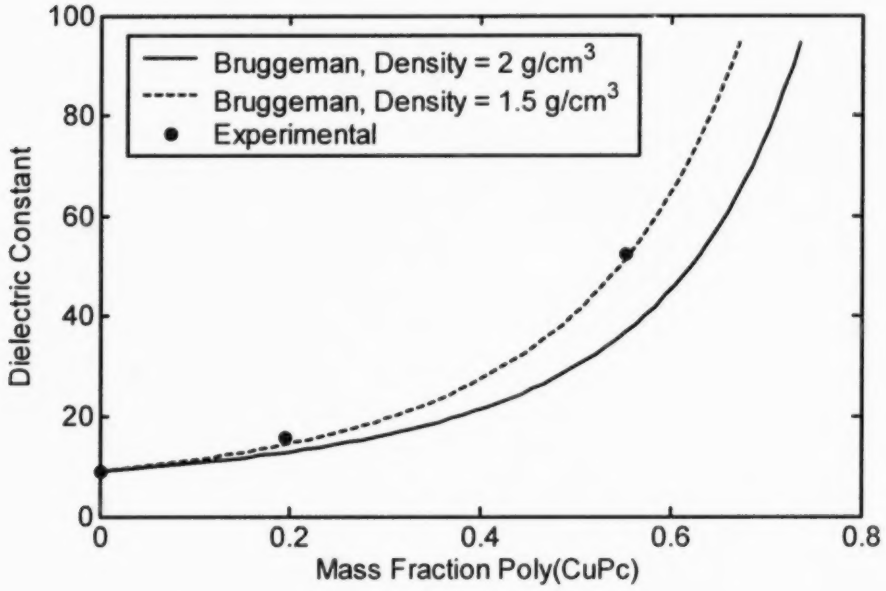


Figure 10. *Bruggeman model of polyurethane/poly(copper phthalocyanine) composite*

$$\frac{\varepsilon_p - \varepsilon_c}{\varepsilon_p - \varepsilon_m} = (1-\phi)\left(\frac{\varepsilon_c}{\varepsilon_m}\right)^{1/3} \tag{7}$$

Comparisons of experimental data with equation (7) are shown in Figure 9 and Figure 10. Experimental values for composites with high $BaTiO_3$ mass fractions could not be collected because the films were too brittle to be removed intact from the release layer. Some uncertainties arose regarding which density to use for the poly(copper phthalocyanine). The supplier did not provide a value, and literature values range from 0.5 – 2.0 g/cm^3. A value of 2.0 g/cm^3 was obtained by determining the density of the polyurethane/poly(CuPc) composite (mass divided by measured volume) and then calculating the density of the poly(CuPc) knowing the density of the polyurethane. This method assumed that the volumes were additive. The best fit occurs using a density of 1.5 g/cm^3. Clearly, more data points would have been helpful, but the number of measurements was limited by poly(CuPc) availability.

Breakdown strength test results for unfilled polymer seem to indicate that breakdown strength is proportional to film thickness (Figure 11). This result is contrary to previous findings wherein breakdown strength varied inversely as a fractional power of film thickness or as the reciprocal of the square root of the thickness[11]. One possible explanation, considering the relatively uncontrolled environment in which the films were prepared, is that a defect or air gap of a particular size would be more likely to cause breakdown in a thinner film than a defect or air gap of the same size in a thicker

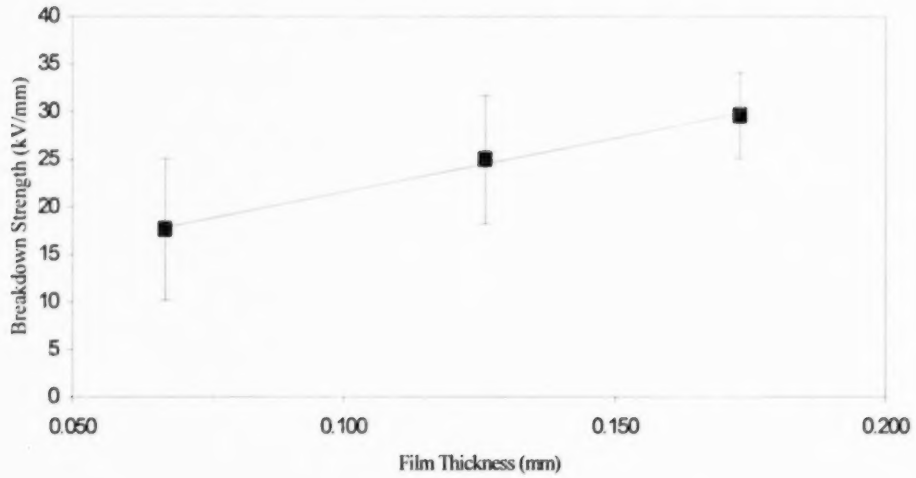


Figure 11. *Breakdown strength as a function of polyurethane film thickness*

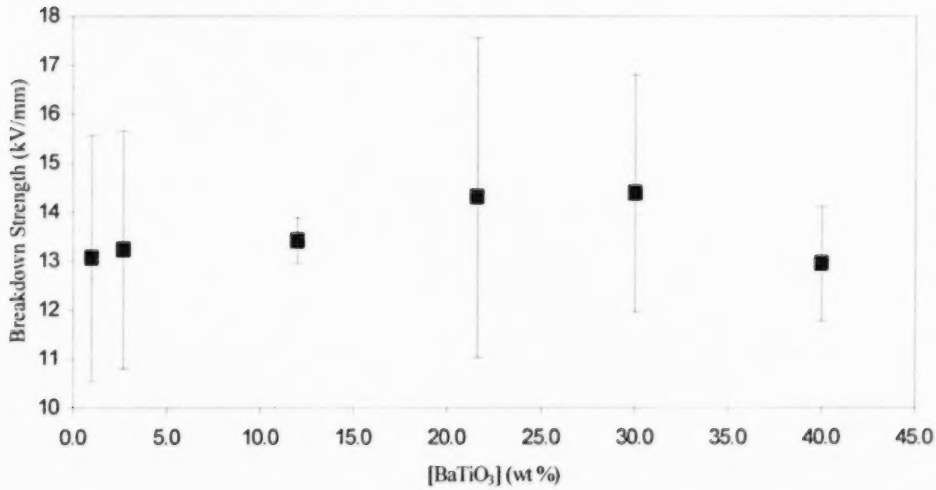


Figure 12. *Breakdown strength as a function of barium titanate concentration (average film thickness = 40 μm)*

film, simply due to the fact that the thicker specimen would provide more insulation around the impurity.

As shown in Figure 12, there is no apparent trend in breakdown strength for composites of varying barium titanate concentration. This is an encouraging result. It demonstrates that a filler can be added to increase a film's permittivity without lowering its breakdown strength. It is important to note that the large error bars are not

Table 3. Breakdown strength variation in a polyurethane/$BaTiO_3$ composite film (1% w/w $BaTiO_3$)

BREAKDOWN STRENGTH (kV/mm)	11.6
	9.7
	13.2
	16.0
	14.8
AVERAGE	13.1
STANDARD DEVIATION	2.5

due to voltage or thickness measurement inaccuracies, but represent true variations in the breakdown strength of the specimen over the area of the film. Because breakdown can occur through numerous mechanisms, it is an extremely variable parameter whose value is difficult to predict with any certainty. Table 3 shows an example of this variation, most of which is due to sample heterogeneity.

5. Conclusions

The thermoplastic polyurethane studied in this work is a poor candidate for the actuating material of a dielectric actuator. Although the polymer shows promising permittivity and dielectric loss, its adhesive-like nature and low strength make it unsuitable for such an application. The dielectric constant of polyurethane/$BaTiO_3$ and polyurethane/poly(CuPc) composites appears to follow the Bruggeman model; more data points, especially in the upper mass fraction range, are needed to confirm this relationship. Barium titanate filler has no significant effect on a film's breakdown strength at mass fractions of less than 0.4.

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Electroactive polymers are an emerging class of actuating materials capable of generating large displacements and fast response times. Dielectric actuators operate on the principle of Maxwell stress, and their response is highly dependent on the material's electrical and mechanical properties. Unfilled and composite films based on a thermoplastic polyurethane were prepared using solvent cast methods, and their dielectric properties were determined. Dielectric constants of up to 52.5 at 100 Hz were measured. The observed permittivities of the composites were consistent with the predictions of the Bruggeman model. Dielectric breakdown strength varied greatly from position to position on each film, reflecting the large statistical variation in flaw density over each film's area. For polyurethane/$BaTiO_3$ composite films, there was no significant change in breakdown strength with increasing barium titanate concentration.

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